MICROWAVE SURFACE RESISTANCE OF Nb FILMS

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Abstract

The surface resistance, R_s, of niobium (Nb) films has been experimentally investigated as a function of thickness, preparation technique and substrate material at 8.86 GHz. Nb films were prepared by either sputtering or evaporation in the thickness range between 0.1 μm and 3.0 μm on either copper (Cu) or sapphire substrate. R_s was determined using a cylindrical TE_{011} mode resonant cavity with one removable end-plate which was utilized as the test substrate. The low field R_s at 4.2 K is lower than that of bulk Nb and shows good agreement with BCS calculation which takes into account the effects of mean free path. The temperature dependence of R_s indicates a normalized film gap parameter, $\Delta(0)/KT_c$, nearly equivalent to the bulk value for most of the films. At low temperatures, R_s is dominated by residual resistance (R_0) which approaches 1 μΩ. The overall characteristics of Nb on Cu (Nb/Cu) indicate that this composite material is potentially useful in applications requiring high rf field as well as high thermal stability.

I. Introduction

At present, only two superconducting materials are utilized in high power microwave applications, Nb and lead (Pb). If properly processed, these two materials maintain low surface resistance up to high microwave surface fields which approach the thermodynamic critical field, H_c. Consequently, resonant cavities fabricated from these superconductors have very low loss and are being used, for example, in the development of nuclear particle accelerators.[1,2]

Nb is technically superior to Pb by approximately a factor of 2 at 10 GHz in terms of the relevant superconducting parameters, H_c and R_s. It is normally utilized in bulk form or in electron-beam welded structures which require relatively elaborate surface electropolish and high temperature vacuum anneal.[3] Pb has been found to require less elaborate surface processing. In fact the theoretical limiting H_c and R_s can be realized on chemically polished, electrodeposited Pb layers only a few microns thick.[4] Thus, superconducting Pb resonators are commonly copper, Cu, structures[5] with only a surface layer of Pb, a few microns thick. The basic Cu structure of these resonators provides relatively great thermal stability. Resonators of this type commonly do not exhibit "run-away" thermal breakdown and consequently multipacting levels are easily processed away.

These preceeding features suggest that thin layers of Nb (or the A-15 compounds) on Cu (Nb/Cu) might form a technically superior microwave superconductor. In order to investigate this possibility, research has been initiated to study the surface resistance and critical field of Nb and A-15 films at microwave frequency. Such a system can be prepared by evaporation, sputtering, or electroplating and has been studied for its dc or very low frequency ac properties.[6,7] Only limited rf or microwave study, however, has been performed on this system in the past.[8] This report is a brief summary of our preliminary results at x-band for Nb films.

II. Experimental Procedure

Films used in this study were produced either by sputtering or evaporation. Substrates were either OFHC Cu or sapphire (of optical window quality supplied by Union Carbide) of disc shape; 6.4 cm in diameter and 3.2 mm in thickness. After machining, Cu substrates were lapped on emery paper of successively finer grit; the final mechanical polish was accomplished with levigated alumina compound of 1-3 μm in particle size. Prior to deposition by either method, Cu discs were electropolished to remove several μm of surface material. Sapphire substrates were cleaned by conventional chemical cleaning agent.

Sputtered samples

Nb films were sputtered onto Cu substrates to the thickness of 0.33, 1.0 and 3.0 μm. Sputtering was performed in a planar diode configuration with target-sample distance of 4.5 cm using an rf power source. First, the substrate was sputter-etched for ~ 1500 Å to remove the surface oxide. Then Nb was sputtered onto the substrate at a typical rate of 300 Å/min in a 10 micron argon atmosphere. During etching and deposition the substrate temperature was estimated to have risen to about 350° C.

Evaporated samples

Nb films were evaporated on Cu substrates at 0.11, 0.33 and 0.80 μm in thickness and on sapphire at 0.33 μm. Substrates were heated to ≃ 400° C in the vacuum chamber (Ultek, all metal, ion pumped) prior to evaporation and maintained near that temperature throughout evaporation. Base pressure was typically below 10^{-8} mm Hg and chamber pressure remained below 2×10^{-7} mm Hg during electron-beam evaporation. Typical average evaporation rate was 300 Å/min.

Microwave measurement

The x-band cylindrical resonator used for this study was made of OFHC copper with a diameter/length ratio of 1.75 and was operated at 8.86 GHz in TE_{011} mode.[15] All measurements were performed in a pulsed mode in which resonator Q, field level and energy content could be determined by measuring the decay time together with the incident, reflected and transmitted powers. For a homogeneous resonator $R_s = \Gamma/Q$ where the geometrical factor Γ is 708 Ω for the present geometry.

For these measurements the entire resonator was first electroplated with Pb to a thickness of 3 μm and relevant parameters such as Q and its field dependence were determined to establish a "baseline" for Pb. Then the bottom plate of the resonator was replaced with a thin film sample plate for a similar set of measurements. The surface resistance of the thin film sample bottom plate, R_1, is related to the measured quality factors by:

$$R_1 = \frac{\Gamma}{Q_0}\left[1 + \frac{1}{f}\left(\frac{Q_0}{Q_1} - 1\right)\right] \qquad (1)$$

where Q_0 is the baseline quality factor of entirely Pb/Cu surface and Q_1 is that of the same cavity with the thin film sample bottom plate. The parameter f is the fraction of the loss arising from the bottom plate

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and is 0.237 for our present geometry.

Transition temperature and resistivity

In order to assess the quality of the sputtered and evaporated films, the transition temperature T_c was determined inductively at low frequency ($\simeq$ 400 Hz) by measuring the coupling between a pair of coils placed within ~ 1 mm of the sample surface. The sample was thermally anchored to a copper block which was placed in a partially evacuated chamber immersed in a helium bath. The temperature of the copper block and the sample was controlled by a heater and was monitored by a germanium thermometer. By this method the T_c of every sample was determined. For evaporated films, small samples were made on sapphire substrates at the same time as the disc samples were produced. The resistivity, ρ, as well as T_c of these small samples were subsequently determined by a 4-point probe measurement in order to deduce the mean free path of the electrons, ℓ, using a standard relationship between ρ and ℓ.[13]

III. Results and Discussion

This report presents data on several niobium films relating microwave surface resistance R_s at 8.86 GHz to various film parameters.

Transition temperature

The films (see Table 1) range in thickness d $\simeq$ .1 - 3 μm. In this range of thickness the proximity effect of the substrate is small, except for the thinnest films. Target material for the films was 0.999 Nb with T_c = 9.25 K. Precision of the temperature measurement is estimated to be ± 0.025 K based on calibration from T_c of high purity Nb and Pb. We found T_c for sputtered films on Cu to be generally somewhat higher than that of the target material, and that it increased with increasing thickness. Typical transition width (10% - 90%) was $\Delta T \simeq \pm$ 0.020 K. By comparison, evaporated Nb films on Cu had slightly lower transition temperatures (by ~ 0.1 K) than sputtered films of equivalent thickness. Transition temperature data is listed in Table 1 for several films. Note that for thin evaporated films of equal thickness, T_c on sapphire is slightly higher than on Cu -- probably reflecting the proximity effect in the thinner films.

The highest T_c observed was for 3 μ Nb sputtered on Cu where T_c = 9.74 K or 0.5 K above that of the target material. The highest temperature for the onset of superconductivity in this film was 9.89 K. This modest increase in T_c is outside the range of experimental uncertainty and we believe it to be real.

Low field surface resistance

For all films and substrates $R_s(T)$ was found to depend somewhat on peak microwave surface field h_s. However, at low fields $R_s(T)$ is independent of h_s. At higher microwave fields R_s and the microwave critical field show a very strong dependence on substrate material as will be published elsewhere.

The temperature dependence of R_s presented here is for peak surface fields $\lesssim$ 0.3 mT which is well within the linear range of R_s for all films.

Figure 1 indicates (as triangles) data on $R_s(T)$ for Pb/Cu as well as typical data for Nb/Cu films (as circles). The Pb/Cu data comes from measurements on a resonator fabricated entirely of Pb/Cu as discussed previously. The Pb/Cu data is compared with a BCS

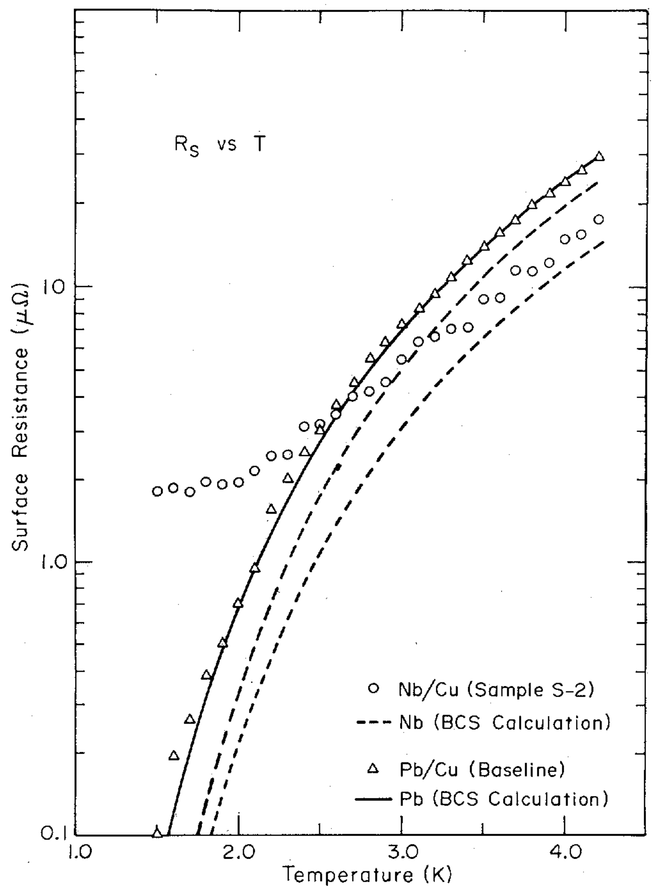

Fig. 1 Surface resistance as a function of temperature. BCS surface resistance is plotted for both Pb and Nb for comparison. Dashed curves (upper: ℓ = 10,000 Å, specular. Lower: ℓ = 300 Å, diffuse).

Preparation Method	Sample ID	Thickness (μm)	Substrate	Disk Sample T_c (°K)	Disk Sample ΔT_c (°K)	Small Sample T_c (°K)	Small Sample ΔT_c (°K)	$\Delta(0)/KT_c$	R_0 (μΩ)	$R_{EXP}(4.2)$ (μΩ)	ℓ (Å)
Sput-tered	S1	3.0	Cu	9.73	.22			1.75	1.55	8.8	
	S2	1.0	Cu	9.49	.04			1.67	1.76	14.5	
	S3	0.33	Cu	9.31	.05			1.63	7.05	14.1	
	S2H	1.0	Cu	8.43	.19			1.77	13.3	31.9	
Evapo-rated	DJ	0.80	Cu	9.27	.04	9.39	.05	1.91	1.28	13.0	103
	DM	0.33	Cu	9.07	.04	9.21	.07	1.69	2.42	15.9	147
	DN	0.11	Cu	8.99	.07	9.22	.03	1.37	45.0	28.8	242
	DL	0.33	Sapphire	9.28	.02	9.28	.02	1.70	2.69	15.2	608-1080

Table 1. T_c = Transition temperature, ΔT_c = Transition width (10-90%), $\Delta(0)/KT_c$ = Normalized gap parameter, R_0 = Residual resistance, $R_{EXP}(4.2)$ = Best fit to surface resistance data at 4.2 K, ℓ = Mean free path.

calculation[9] as the solid curve and shows very good agreement over this entire temperature range ($0.2 < T/T_c < 0.6$) with little residual resistance (excess resistance above BCS) at the lowest temperature. At the lowest temperature ($\simeq 1.5$ K) Pb/Cu surfaces typically showed a residual resistance less than 0.1 μΩ. The Nb data is from measurements on the same resonator with a Nb film endplate, as analyzed in Section II. The dashed curves are the BCS calculated values for R_s utilizing the accepted materials parameters for Nb.[11] The difference between the two curves is due to mean free path effects (discussed later) and represent the extreme limits on R_s due to this effect. In general these Nb films show the ideal BCS surface resistance at the highest temperature ($T \sim 4.2$ K, $T/T_c \sim 0.45$) but a relatively large residual resistance at lower temperature.

Further comparison between Nb film data and the BCS calculation was made by performing a least-squares fit to the following equation:[10]

$$R_s(T) \;=\; A \exp\left(-\frac{\Delta(0)}{KT_c}\,\frac{T_c}{T}\right) + R_0 \qquad (2)$$

where the first term represents an approximation to the BCS surface resistance below $T \leq 0.5\,T_c$ and the second term accounts for a temperature insensitive residual resistance, R_0. The parameters utilized in fitting this equation to the data were A, zero temperature gap $\Delta(0)$ and residual resistance R_0. Figure 2 illustrates surface resistance data fit in this way but R_s is now plotted against T_c/T. Raw data are

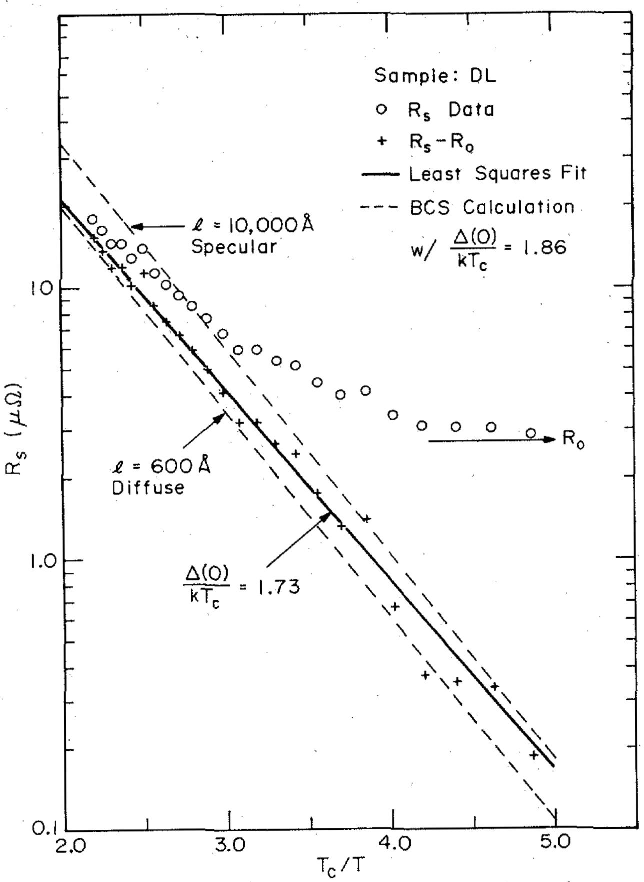

Fig. 2 Surface resistance for Nb films plotted against the reciprocal of normalized temperature. Dashed lines are BCS calculations (see text).

plotted on circles while the crosses represent the raw data minus a residual resistance factor. The solid line is a least square fit evaluating $\Delta(0)$. In Table 1 these parameters are listed for several films. Also listed as $R_{EXP}(4.2)$ is the best fit for $R_s - R_0$ from Equation (2). These quantities are discussed below.

Gap parameter. For bulk Nb cavities, previous microwave measurements of $R_s(T)$ indicate a normalized gap parameter, $\Delta(0)/KT_c \simeq 1.85$;[10] previous tunneling data on Nb films gives $\Delta(0)/KT_c \sim 1.9$.[12] As shown in Table 1, our data, utilizing R_s from these Nb/Cu films, leads to a normalized gap parameter ~ 1.8 which is a slight function of thickness and is generally larger for the films with lower R_0.

Ideal surface resistance. The BCS surface resistance can be extracted from Equation (2) as $R_s(T) - R_0$. This quantity has been calculated[10] and is a function of mean free path ℓ. The theory predicts a minimum in $R_s(T) - R_0$ when ℓ is about equal to the London penetration depth, λ_L, which is $\simeq 330$ Å for Nb. This minimum becomes more pronounced at low frequency. The calculated[9] dependence of $R_s(T) - R_0$ on ℓ for Nb at 4.2 K and at 8.86 GHz is shown as curves in Figure 3, using standard material parameters.[11] Also indicated are experimental evaluations of this quantity, R_{EXP} (see Table 1), shown as a function of mean free path for several evaporated films. For these films the mean free path was deduced from the measured resistivity, just above T_c, of small samples on sapphire substrates produced during the same evaporation. Except for the thinnest (.11 μm) film the experimental values for R_{EXP} are consistent with the theoretical calculation. Note that this resistance is less than for bulk Nb which typically has a larger ℓ than the present films. For the thinnest film we suspect only partial screening of the surface current by the film as will be discussed later.

For sputtered films, experimental values for mean free path were not available; consequently R_{EXP} data are indicated only at the right side of Figure 4 as

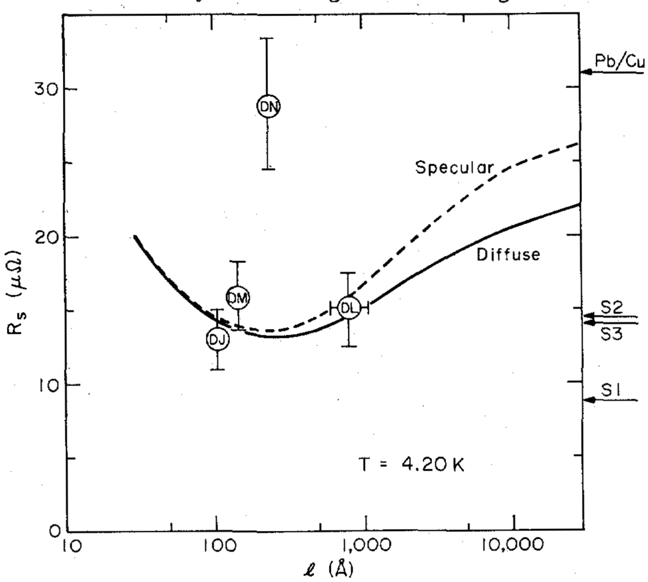

Fig. 3 Surface resistance of Nb films at 4.2 K as a function of electronic mean free path and surface reflection.

S1, S2, and S3 (see Table 1). These data imply a short mean free path: S2 and S3 indicate that $100 \leq \ell \leq 1000$ Å. The thick sputtered film, S1, was found to have anomalously small R_{EXP} which is $\simeq 30\%$ below the minimum value given by the calculation; this film also has the highest transition temperature (see Table 1).

Residual resistance. For these films R_0 is typically a few micro-ohms. The residual resistance increases as thickness decreases and for equivalent thickness is lower for the evaporated films. The thinnest evaporated film (d = .11 μ) is only approximately two penetration depths thick and the relatively large R_0 is due to flux leakage into the substrate.

A photomicrograph of a typical 0.8 μ Nb film (DJ in Table 1) on Cu substrate is shown in Figure 4(a) where the line structures are the grain boundaries of the Cu substrate. Small pits, also shown in Figure 4 (a) are typically a few μm in diameter, and are predominantly located along grain boundaries. In order to determine the uniformity of Nb coverage, random areas of several samples were scanned under SEM searching for x-ray emission from Cu. The result showed no discontinuity in the Nb coverage across grain boundaries and in general Nb coverage was complete even for the thinnest sputtered and evaporated films.

On the other hand some of the pits such as the one shown in the middle of Figure 4(b) showed an excessive amount of Cu indicating that these pits were not entirely covered with Nb. This type of imperfection can easily give rise to R_0 via normal metal dissipation. We estimate that such normal spots, if present at a fractional level of $\sim 2 \times 10^{-4}$, would lead to $R_0 \sim 1$ μΩ (the normal state surface resistance of Cu was measured to be 5.6 mΩ). Thus at this grain density, only one pit in ten grains could account for the observed R_0. Generally R_0 decreases as the thickness is increased for both preparation methods, which seems to indicate that the Nb coverage improves with thickness.

An attempt was made to anneal one of the samples (S2) above the stress annealing temperature (750° C) of Nb. It was annealed at 800° C for ~ 3 hours in a vacuum oven at < 5×10^{-6} mm Hg. The heat treatment resulted in a marked decrease in T_c of the film from 9.49 to 8.43 K and at the same time considerable increase in R_0 as well as $R_{EXP}(4.2)$, as entered in Table 1 by S2H. One possible cause of this behavior is the partial diffusion of Cu into Nb which reduces the mean free path to increase the BCS part of surface resistance (to the left side of Figure 4) and at the same time increases the effective penetration depth so that the surface current leaks into the substrate resulting in an increased R_0.

When the film thickness becomes comparable to effective penetration depth (400 - 800 Å for these Nb films), because of field leakage into the substrate Cu, the surface resistance becomes a function of thickness. It is possible to estimate the approximate magnitude of this effect by calculating the effective penetration depth of Nb for a known mean free path using the same computer program described earlier. The results show that except for the very thinnest film used (sample DN), the current leakage should give no more than 0.2 μΩ in R_0. However for the thinnest film sample DN, these calculations indicate that a major fraction of R_0 (45 μΩ) is due to this effect.

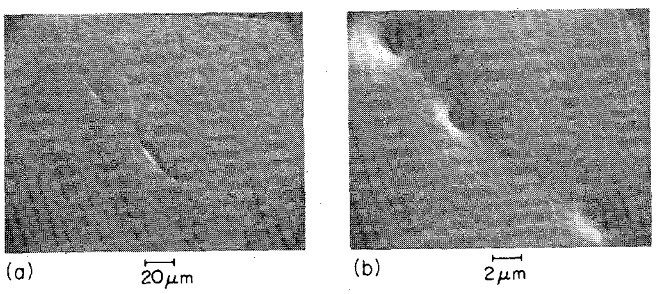

Fig. 4 Photomicrograph of (a) sample DJ, (b) pits along grain boundaries.

Finally it should also be pointed out that there are some variables that depend on the processing method since two films of the same thickness produced by sputtering versus evaporation give significantly different R_0 (7.05 μΩ for S3 as opposed to 2.42 μΩ for DM).

A-15 Compound

A natural extension of this technique is to superconducting films of A-15 compounds on Cu substrates. Preliminary investigation has been initiated in this direction by forming Nb_3Au compound in two ways. One method is by evaporating a layer of Au (typically 2700 Å) between two Nb layers (typically 3500 Å) on Cu substrate and subsequently heat treating at ~ 700° C in vacuum for several hours. Another method is by sputtering a 1 μm layer of Au on bulk Nb followed by a similar heat treatment. Films thus produced show transition temperatures of 10.79 K and 10.89 K, respectively, which are close to the published T_c (= 11.0 K) of bulk Nb_3Au.[14] Microwave measurement and surface analysis of films thus produced are underway.

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